Nano Polyurethane (Nano PU) was used for the fabrication of multilayer nanocomposite film deposition on wool fabrics by electrostatic self-assembly to improve the anti-felting properties. Oppositely charged cationic poly (diallyldimethylammonium chloride) (PDDA) and anionic Nano PU were alternately deposited on the surface of wool fabrics. 8, 12 and 16 multilayer films of PDDA/Nano PU were deposited on the wool fabric surfaces using a padder. Attenuated Total Reflectance Fourier Transform Infrared Spectroscopy (FTIR-ATR) and Scanning Electron Microscopy (SEM-EDX) were used to verify the presence of deposit nanolayers. Breaking strength, whiteness and yellowness value analysis was performed on the fabrics before and after the treatment with Nano PU by the electrostatic self-assembly method. The build-up of the multilayer films and the level of colour strength (K/S) achieved are discussed after the acid dyeing process. To examine the anti-felting properties of the multilayered fabrics, the fabric shrinkage after washing was determined.